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ABSORPTION OF LIGHT AND HEAT RADIATION BY SMALL SPHERICAL PARTICLES

II. SCATTERING OF LIGHT BY SMALL CARBON SPHERES[1]

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Abstract

Spheres of carbon for which $2a/\lambda$, the ratio between the diameter of the particle and the incident wave-length, is less than about $\frac{1}{10}$ scatter the light uniformly in all directions. The intensity of the scattered radiation for any angle is proportional to the square of the volume of the particle and inversely proportional to the fourth power of the wave-length. As the ratio $2a/\lambda$ increases from $\frac{1}{4}$ to $\frac{1}{2}$ and greater values, the diffused light collects more and more into a main beam that appears as a continuation of the incident ray and that decreases in width as $2a/\lambda$ increases. Blue light prevails in the scattered radiation. When the size of the particles is unknown, the intensity, distribution, and polarization of the scattered light give an at least approximate value for the radius.

Amount of Light Scattered by Small Spherical Particles of Carbon

The loss of light observed behind a small spherical obstacle brought into the path of a plane wave of light may be large for two reasons, either because a large portion of the incident light that enters the obstacle is transformed into heat, or because a large portion of the incident light is deflected in other directions by the obstacle. It is to be expected that if the sphere consists of carbon, absorption accounts for the larger part of the loss; indeed, the relatively high index of refraction of carbon indicates that the electrons yield readily to an external alternating field; the conductivity, however, is moderate and the ohmic losses caused by the motion of the electrons are considerable.

In order to determine exactly what fraction of the total loss measured behind a spherical carbon particle is due merely to scattering or diffraction of the light, and what fraction is lost by true absorption, it is necessary to consider both the real and the imaginary part of the amplitudes a_ν and p_ν of the component electric and magnetic waves for which the solution of the problem is obtained. For an incident wave of unit intensity in air these amplitudes are as follows.

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$$a_\nu = \frac{(-1)^\nu(2\nu+1)}{-i+(-1)^\nu \dfrac{J_{-\nu-\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)} \dfrac{\dfrac{\beta}{\alpha}\dfrac{J'_{-\nu-\frac{1}{2}}(\alpha)}{J_{-\nu-\frac{1}{2}}(\alpha)} - \dfrac{J'_{\nu+\frac{1}{2}}(\beta)}{J_{\nu+\frac{1}{2}}(\beta)}}{\dfrac{\beta}{\alpha}\dfrac{J'_{\nu+\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)} - \dfrac{J'_{\nu+\frac{1}{2}}(\beta)}{J_{\nu+\frac{1}{2}}(\beta)}}}$$

$$p_\nu = \frac{(-1)^{\nu+1}(2\nu+1)}{-i+(-1)^\nu \dfrac{J_{-\nu-\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)} \dfrac{\dfrac{\beta}{\alpha}\dfrac{J'_{\nu+\frac{1}{2}}(\beta)}{J_{\nu+\frac{1}{2}}(\beta)} - \dfrac{J'_{-\nu-\frac{1}{2}}(\alpha)}{J_{-\nu-\frac{1}{2}}(\alpha)}}{\dfrac{\beta}{\alpha}\dfrac{J'_{\nu+\frac{1}{2}}(\beta)}{J_{\nu+\frac{1}{2}}(\beta)} - \dfrac{J'_{\nu+\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)}}}.$$

In the present instance, when $\alpha = 2\pi a/\lambda$ the derivatives

$$J'_{\nu+\frac{1}{2}}(\alpha) = J_{\nu-\frac{1}{2}}(\alpha) - \frac{\nu}{\alpha} J_{\nu+\frac{1}{2}}(\alpha)$$

$$J'_{-\nu-\frac{1}{2}}(\alpha) = -J_{-\nu+\frac{1}{2}}(\alpha) - \frac{\nu}{\alpha} J_{-\nu-\frac{1}{2}}(\alpha),$$

are approximately equal to the derivatives of the Bessel functions of half-integral order. The real part and the imaginary part of each of the components a_ν and p_ν are entirely different functions of the ratio $2a/\lambda$ or of $\alpha = 2\pi a/\lambda$, as a glance at Figs. 1 to 4 will show. For very small values of α the value of a_ν increases as $\alpha^{2\nu+1}$; that for p_ν increases as $\alpha^{2\nu+3}$. For larger and larger values of α, the rate of increase of a_ν and p_ν diminishes, and after a few fluctuations, a_ν and p_ν vary regularly according to the law

$$a_\nu = \frac{2\nu+1}{n-in\kappa+1} i^\nu(\cos\alpha + i\sin\alpha)\left((n-in\kappa)i\cos\left(\alpha-\nu\frac{\pi}{2}\right)+\sin\left(\alpha-\frac{\nu\pi}{2}\right)\right)$$

$$-p_\nu = \frac{2\nu+1}{n+1-in\kappa} i^\nu(\cos\alpha+i\sin\alpha)\left(i\cos\left(\alpha-\nu\frac{\pi}{2}\right)+(n-in\kappa)\sin\left(\alpha-\frac{\nu\pi}{2}\right)\right).$$

It follows that in this final stage the real parts of a_ν and p_ν oscillate between

$$\pm\frac{2\nu+1}{2}\,\frac{1-n^2-n^2\kappa^2}{(1+n)^2+n^2\kappa^2},$$

the positive values occurring for multiples of π, the negative for odd multiples of $\pi/2$ when the formula is used for a_ν (Figs. 2 and 4). For surroundings other than air, n has to be divided by the refractive index m_0 of the medium.

The imaginary portions of a_ν and p_ν, on the other hand, reach much higher values (Figs. 1 and 3); apart from the sign, which remains unchanged, these values alternate regularly between

$$(2\nu+1)\frac{1+n}{(1+n)^2+n^2\kappa^2} \text{ and } (2\nu+1)\frac{n^2+n+n^2\kappa^2}{(1+n)^2+n^2\kappa^2},$$

and are thus simply proportional to $(2\nu+1)$.

When, therefore, larger and larger ratios are chosen for α, the imaginary part of the difference $(-1)^\nu(a_\nu - p_\nu)$, which determines the total loss of light along the direction of the incident ray, becomes equal to $i\nu^2$. Calculations show the number ν of components that contribute appreciably to

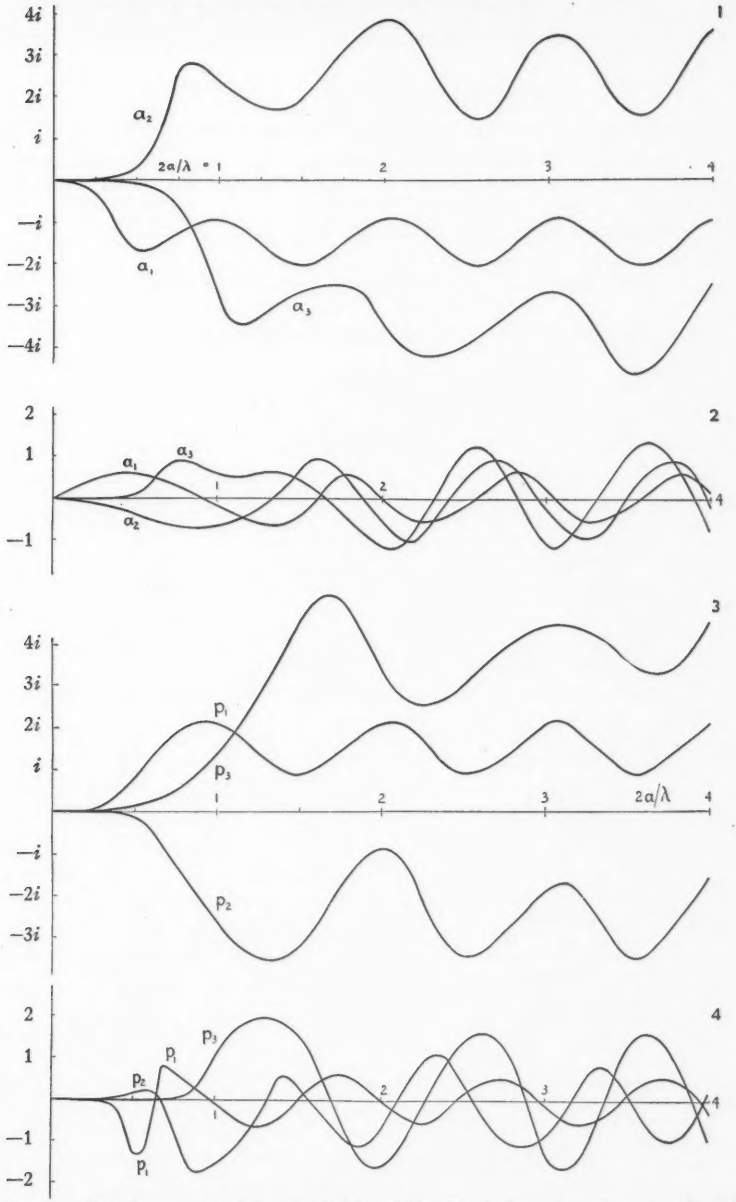


FIG. 1. *Imaginary part of the amplitudes of the electrical component waves set up by spherical carbon particles of different ratios* $2a/\lambda$. FIG. 2. *Real part of the amplitudes of the electrical component waves set up by the particles.* FIG. 3. *Imaginary part of the amplitudes of the magnetic component waves sent out by particles having different ratios* $2a/\lambda$. FIG. 4. *Real part of the amplitudes of the component magnetic waves sent out by carbon particles.*

the sum of all the expressions $(-1)^{\nu}(a_{\nu} - p_{\nu})$ to be equal to α; there follows the relation

$$\text{imag. part of } \Sigma(-1)^{\nu}(a_{\nu} - p_{\nu}) = \alpha^2 .$$

Hence the actual surface, $2\pi a^2$, that the obstacle presents to the light, and not merely its cross-section, is involved in the absorption and deflection of the wave.

Of the fraction lost per unit length of path,

$$k = \text{imaginary component of } \frac{\lambda^2}{2\pi m_0^2}\Sigma(-1)^{\nu}(a_{\nu} - p_{\nu}),$$

when there is one particle per unit volume, the amount

$$k_s = \frac{\lambda^2}{2\pi m_0^2}\,\Sigma\,\frac{|a_{\nu}|^2 + |p_{\nu}|^2}{2\nu + 1}$$

is lost as a result of the deflection of the light in other directions. The symbol $|a|$ means the absolute value of the complex quantity a. The values calculated for carbon particles are shown in Fig. 5 for various diameters of the

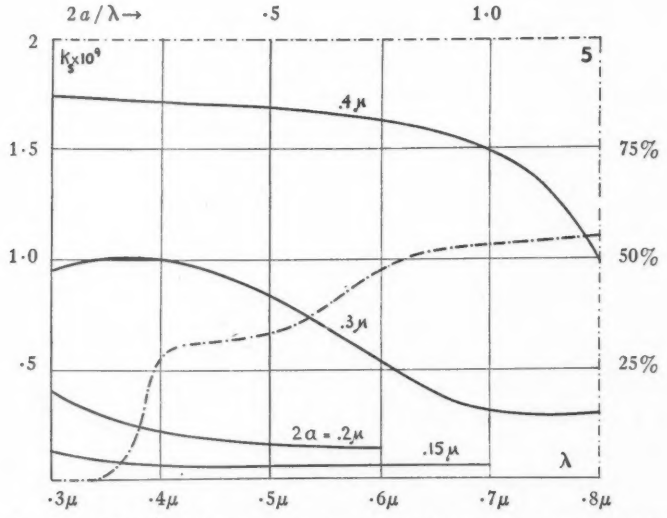


FIG. 5. *Loss of light caused by scattering for one particle per unit volume and percentage of total loss caused by diffusion (dot-dash lines).*

particle ($2a = 0.15$, 0.2, 0.3, and 0.4 μ). They are seen to be small in the visible region at least, and to decrease in general with the wave-length, most steeply when the diameter $2a$ of the particle is in the neighbourhood of 0.3μ; for this size the loss in the visible diminishes markedly as the wave-length is increased.

Ratio Between Loss by Deflection and Total Loss

For very small values of α, only one component, the electric wave of amplitude a_1, where

$$a_1 = 2\alpha^3 \frac{(n - in\kappa)^2 - 1}{(n - in\kappa)^2 + 2},$$

need be considered. It follows that the ratio of deflected light to the total fraction of the radiation lost along the direction of the incoming wave, owing to the presence of the particle, is equal to

$$R = 2\alpha^3 \frac{(n^2 - n^2\kappa^2 - 1)(n^2 - n^2\kappa^2 + 2) - i6n\kappa}{(n^2 - n^2\kappa^2 + 2)^2 + 4n^2\kappa^2}.$$

The ratio is zero when the particles are extremely small but increases considerably as values between $\pi/10$ and $\pi/4$ are given to α. For very large values of α the fraction tends towards the limit

$$R_l = \frac{(n^2 + n + n^2\kappa^2)^2 + (1 + n)^2 + 2n^2\kappa^2}{((1 + n)^2 + n^2\kappa^2)^2},$$

or towards about 0.6 for carbon particles. Fig. 5 shows the changes in the proportion of diffracted or scattered light for carbon particles of various sizes.

Angular Distribution of the Deflected Light

The amount of light that is deflected by a spherical obstacle depends on the angle between the direction considered and the direction of the incident wave. The variation of the intensity with the angle merits examination on account of its close relation to the size of the particle.

When a spherical particle is placed in the path of a plane unpolarized wave of strong monochromatic light, the intensity of the scattered light observed in a direction that forms the arbitrary angle γ with the incident beam sent along the horizontal or z-axis is obtained by the simple addition of the two polarized components $I_\perp$ and $I_\parallel$. The intensity $I_\perp$ is the intensity of the light whose electrical vibration is perpendicular to the plane containing the line of vision and the incident ray, and $I_\parallel$ is the intensity of the light whose electric vector oscillates in the plane determined by the line of vision and the incident ray. When the distance r separating observer and particle is a large multiple of a, the general formulae for the intensity $I_\perp$ and $I_\parallel$ are

$$I_\perp = \frac{1}{\rho^2}\left|\sum_1^\infty \left(\frac{a_\nu}{\nu(\nu+1)}\,\Pi_\nu + \frac{b_\nu}{\nu(\nu+1)}\,(v\Pi_\nu - (1 - v^2)\Pi'_\nu)\right)\right|^2$$

$$I_\parallel = \frac{1}{\rho^2}\left|\sum_1^\infty \left(\frac{a_\nu}{\nu(\nu+1)}\,(v\Pi_\nu - (1 - v^2)\Pi'_\nu) + \frac{b_\nu}{\nu(\nu+1)}\,\Pi_\nu\right)\right|^2,$$

where

$$v = \cos\gamma$$

$$\rho = \frac{2\pi r}{\lambda} m_0$$

$$\Pi_\nu = \frac{1}{2^\nu} \sum_0^{\frac{\nu+1}{2}-1} (-1)^s \frac{(2\nu - 2s)!\, v^{\nu-2s-1}}{(\nu - s)!\, s!(\nu - 2s - 1)!}.$$

When the upper limit of $s = (\nu + 1)/2$ is half an integer, this particular value must not be considered in evaluating Π_ν.

The sums $(I_\perp + I_\parallel)$ to be expected at a given large distance from carbon particles of different diameters have been calculated, and are represented in Figs. 6 to 9 along the directions for which they are valid; the small spheres are assumed to be of such size that when illuminated with light of a constant wave-length the ratio $2a/\lambda$ is a multiple of $\frac{1}{8}$. Grains for which $2a/\lambda$ is less than $\frac{1}{8}$ are seen to diffuse light uniformly in all directions. For ratios from $\frac{1}{4}$ upwards, light is deflected only slightly from the direction of the incoming ray. For $2a/\lambda = \frac{1}{2}$, for instance, by far the larger part of the light coming from the particle is confined to the space within the angles 120° and 240°. With still larger spheres (Fig. 9) only a narrow main beam, accompanied by a few unimportant side lobes, remains and very little light is visible at right angles to the direction of the incoming wave.

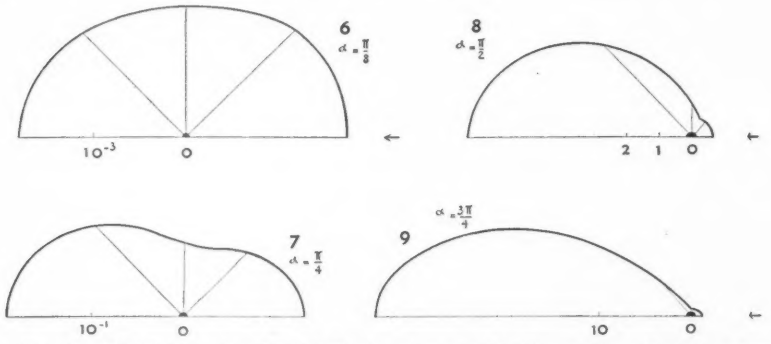


FIG. 6. *Distribution of scattered light of a given wave-length around a spherical carbon particle for which* $2a/\lambda = \frac{1}{8}$. *The light is coming from the right.*

FIG. 7. *Distribution of scattered light around a spherical carbon particle for which* $2a/\lambda = \frac{1}{4}$.

FIG. 8. *Distribution of scattered light around a spherical carbon particle for which* $2a/\lambda = \frac{1}{2}$.

FIG. 9. *Distribution of scattered light around a spherical carbon particle for which* $2a/\lambda = \frac{3}{4}$. *Scale units are 10,000 times as large as those used for* $2a/\lambda = \frac{1}{8}$ *and 100 times as large as those used for* $2a/\lambda = \frac{1}{4}$.

As regards the intensity of the radiation at a given distance, only a small amount of light is diffused by particles having a diameter less than one-eighth of the wave-length of the incident light. For $2a/\lambda = \frac{1}{8}$, the light contributed by 10,000 particles is required to equal the intensity obtained in any direction from a sphere for which $2a/\lambda = \frac{3}{4}$. According to the formula valid for the smallest particles,

$$I_\perp = \frac{\lambda^2}{16\pi^2 r^2} |a_1|^2$$

in any direction. When the volume $V = 4\pi a^3/3$ is introduced, this expression can be written

$$I_\perp = \frac{9V^2}{4r^2\lambda^4} \frac{|a_1|^2}{\alpha^6} = \frac{9V^2}{r^2\lambda^4} \left| \frac{(n - in\kappa)^2 - 1}{(n - in\kappa)^2 + 2} \right|^2,$$

whereas I_{II} is negligible. That is to say, the intensity of the scattered light in any direction at a distance r from a carbon sphere for which $2a/\lambda$ is less than about $\frac{1}{10}$ is proportional to the sixth power of the radius a and inversely proportional to the fourth power of the wave-length. A steep increase in the intensity of scattered light occurs therefore with increase in diameter. But for larger and larger obstacles, the outgoing light becomes concentrated along the direction of the incoming ray; the intensity of this light varies more nearly as the volume or mass of the particle of carbon, or even less steeply. When, for instance, the intensities obtained behind a spherical obstacle for which $2a/\lambda = 0.5$ are multiplied by $3\frac{3}{8}$ (the ratio between the volumes or masses of spheres for which $2a/\lambda = 0.5$, in one case, and $2a/\lambda = 0.75$ in the other), the results obtained exceed the values given by the larger particles, or are at least of the same order of magnitude (Figs. 8 and 9).

Polarization of Scattered Light

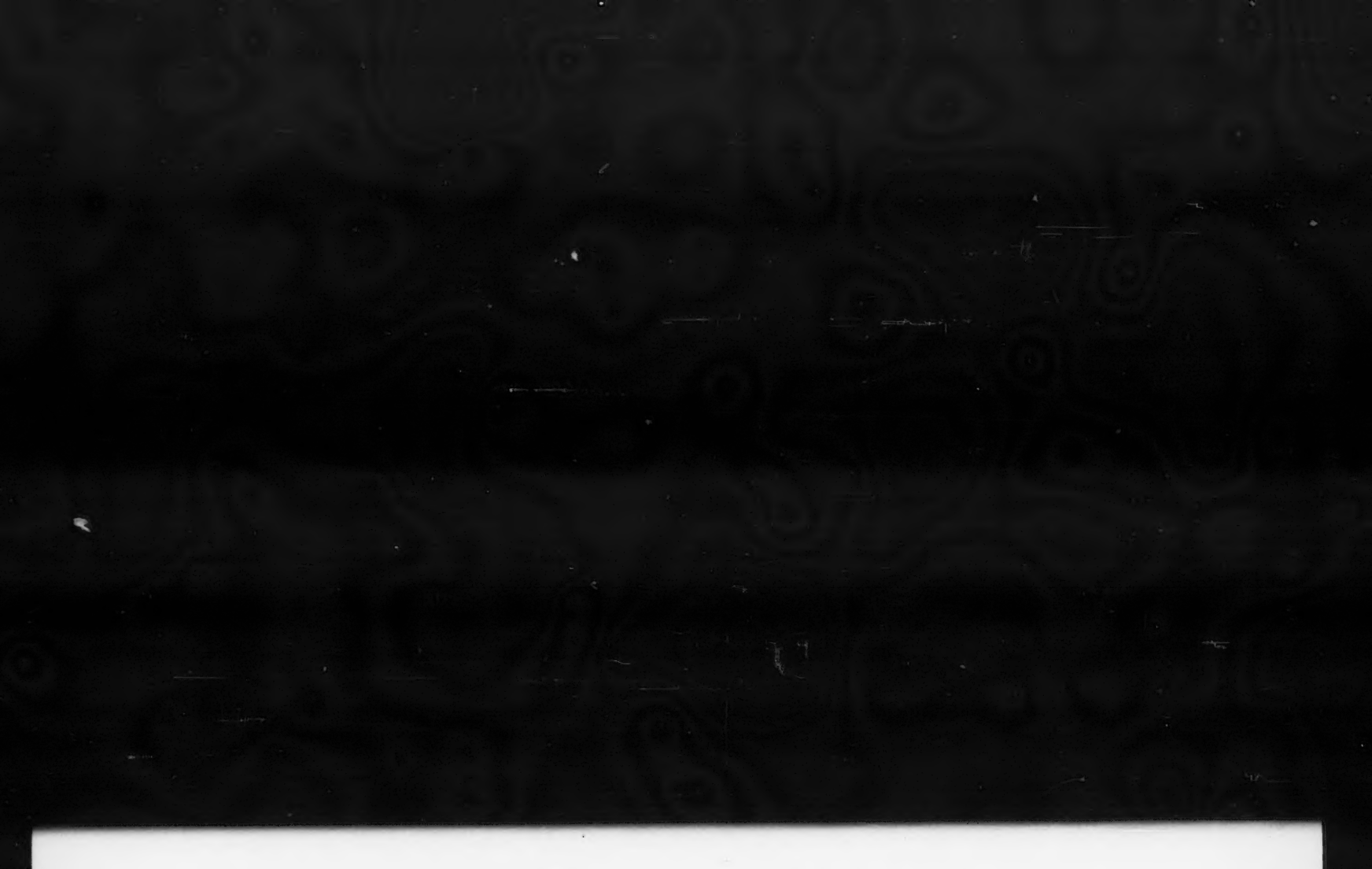
In the light deflected and scattered by the particle along a given horizontal direction, the intensity furnished by the vibration that occurs parallel to the vertical or x-axis is no longer equal to the intensity of the vibration in the horizontal plane. There is therefore in the scattered light an excess $I_I - I_{II}$ or a fraction $(I_I - I_{II})/(I_I + I_{II})$ of polarized light. The vibrations are parallel to the vertical axis when the fraction is positive, and parallel to the plane passing through the incident ray and the direction of vision when it is negative. The fraction varies with the angle γ and the size of the particles. For light scattered by molecules, the radiation emitted in a direction that forms 90° with the incoming ray is completely polarized, the electrical vibrations occurring in the direction that is normal to the plane passing through the incoming and the outgoing rays. The same result is obtained with small spherical carbon particles, as long as the ratio $2a/\lambda$ is less than about $\frac{1}{4}$. When this fraction exceeds $\frac{1}{4}$, the polarization is never quite complete whatever the direction of vision, but the percentage of polarized light attains a maximum for a certain angle or for several angles. For $2a/\lambda = \frac{1}{2}$, the maximum is equal to about $\frac{3}{8}$ near 90°; for $2a/\lambda = \frac{3}{4}$ there occurs almost complete polarization for an angle of 60°, where however the intensity of scattered light is very weak; for directions between 80° and 110° the electrical vibrations in the very faint scattered radiation are parallel to the plane containing the incoming ray and the direction of vision. At 120° the extent of polarization passes through a second peak of about 50%. This variation is a useful indication of the size of the particles.

Practical Applications

The light transmitted or scattered by a single grain of carbon is too weak to be studied; but if a small cloud of particles of similar sizes is contained within a small space, the angles between the incoming and outgoing rays remain virtually unchanged for a distant observer, whatever the particle

chosen, and the formulae remain valid for the total radiation received from the accumulation of N particles.

The most straightforward application of the theory consists of the calculation of the optical properties that a known number of carbon particles of diameter $2a$ suspended in unit volume of air would possess either in white or in monochromatic light.

In practice the reverse problem may also occur; when absorption and scattering per unit volume have been determined experimentally, the concentration of the particles and their size, if approximately uniform, have to be found. If it is possible to measure the relative amount of scattered light at a few angles and for one state of polarization, and if the intensities obtained are plotted as a function of the angles, a comparison of the curve with the theoretical curves for different values of $2a/\lambda$ (Figs. 6 to 9, for instance) suggests an approximate value for the diameter of the particle. A uniform distribution of the diffused light is proof of a diameter less than about $\lambda/10$; absence or weakness of scattered light along directions that form acute angles with the direction of the incident wave points to the presence of coarse carbon sphericles for which $2a/\lambda$ is greater than $\frac{1}{2}$.

As soon as the size is known, the coefficient of absorption can be computed for one particle in unit volume, and the number of particles N can be deduced from the measured absorption per unit volume of suspension. The result arrived at for the concentration of the spheres can be verified by measuring the actual intensity of the diffused light, expressed as a fraction of the incident light, along at least one direction of observation, and by comparing it with the calculated intensity for N grains of carbon.